

# Interference of Instanton Trajectories in Quantum Tunneling for Small Particles of Real Antiferromagnets

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For a two-sublattice antiferromagnet the Lagrangian is constructed taking into account Berry phase whose form is matched with the quantum-mechanical Heisenberg Hamiltonian. Tunnel effects are analyzed taking into account the crystallographic symmetry and possible types of Dzyaloshinskii-Moriya interaction. It is shown that, when the real magnetic symmetry and the Dzyaloshinskii-Moriya interaction are taken into consideration, the effects of a destructive instanton interference and the suppression of macroscopic quantum tunneling can play an essential role. It also may lead to a periodic dependence of the ground-state level splitting on the Dzyaloshinskii-Moriya interaction constant; the magnitude of this splitting is calculated.

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## I. INTRODUCTION

During the last decade, macroscopic quantum tunneling in macroscopic (or, to be more precise, mesoscopic) magnetic systems has become an object of intense experimental and theoretical investigations.<sup>1</sup> In the physics of magnetism such systems include small magnetic particles, magnetic clusters, and high-spin molecules. Special attention is paid to the coherent macroscopic quantum tunneling (CMQT) between physically different, but energetically equivalent, states in systems with discrete degeneracy of the ground state. A typical CMQT effect in such systems is tunneling between two equivalent classical states corresponding to two minima of the anisotropy energy.<sup>2</sup>

The CMQT effects can be observed experimentally from the resonant absorption of electromagnetic waves at tunnel-split energy levels. The interest in these effects are associated with the two following factors. First, mesoscopic objects exhibiting quantum-mechanical properties are interesting as potential elements for quantum computers. Second, fine and elegant effects of interference of instanton trajectories emerge in these problems. For ferromagnetic particles these effects suppress tunneling for half-integral values of the total spin of the system<sup>3,4</sup> and lead to oscillatory dependences of the tunnel splitting of energy levels on extrinsic parameters.<sup>5</sup> In addition, in contrast to the effects of quantum escape from a metastable to stable state the manifestations of the CMQT effects are not masked by thermal fluctuations.

Initially, the CMQT investigations were carried out for small particles of a ferromagnet<sup>6,7</sup> under the assumption that all spins in the particle are parallel to one another (the high-spin model). The effects of destructive interference of instanton trajectories and interference suppression of tunneling were predicted precisely for such systems.<sup>3,4,8</sup> It turned out later that antiferromagnets form a more convenient class for experimental investigations of CMQT. According to calculations of Refs.<sup>9,10</sup>, the level splitting in antiferromagnets is stronger than in

ferromagnets, and the effects can be observed at high temperatures. It is not surprising that the CMQT effects were observed for the first time in ferritin particles with an antiferromagnetic structure.<sup>8</sup> No interference effects are observed in pure antiferromagnets (i.e., in the case of complete compensation of the spins of sublattices), but such effects may appear in the applied magnetic field.<sup>5</sup> It will be shown below that even at zero field the interference effects can also appear when the real magnetic symmetry of the crystal is taken into account, in particular, in the presence of the Dzyaloshinskii-Moriya (DM) interaction.

A semiclassical description of magnetic systems is based on the formalism of coherent spin states. In order to construct the effective field Lagrangian both for ferromagnets and antiferromagnets, we will proceed from the expression for the Euclidean Lagrangian of an individual spin, which has the form<sup>11</sup>

$$\mathcal{L}_0 = -i\hbar s \sum_k \dot{\phi}_k (1 - \cos \theta_k) + W(\phi_k, \theta_k). \quad (1)$$

Here,  $s$  is the spin associated with each magnetic moment,  $\phi_k$  and  $\theta_k$  are the polar coordinates of the  $k$ th magnetic moment, and  $W(\phi_k, \theta_k)$  is the classical magnetic energy of the magnet; the overdot indicates the differentiation with respect to the imaginary time  $\tau = it$ . The first term determines the magnetization dynamics (its variation leads to the well-known Landau-Lifshitz equations in the angular parametrization) and also determines a so-called Berry phase.<sup>11,12</sup> This quantity is associated with the total time derivative which is not manifested itself in the equations of motion, but it is responsible for the interference of instanton trajectories.

For a macroscopic description it is natural to use one or several field variables (order parameters) instead of the set of microscopic variables. The determination of the number of order parameters and their transformation properties in magnetic systems is a nontrivial problem. In the approach based on coherent spin states the order parameter for ferromagnets is the magnetization vector of a constant length, which can be parametrized by the

angular variables  $\theta$  and  $\phi$ . In this case the Berry phase is just a resultant change in the angle along the instanton trajectory.<sup>3,4</sup> The behavior of antiferromagnetic systems can be correctly described using a three-component vector of a fixed length, viz., the antiferromagnetism vector  $\mathbf{l}$ , see Refs.<sup>13–15</sup>. The total spin in this case is a slave variable, and it is determined by the vector  $\mathbf{l}$  and its time derivative  $\partial\mathbf{l}/\partial t$ . Dynamic equations for the antiferromagnetism vector  $\mathbf{l}$  can be either constructed from the symmetry considerations<sup>14</sup> or derived from the Landau-Lifshitz equations for the sublattice magnetizations.<sup>16,17</sup> In both these approaches the same classical equations of motion for the unit vector  $\mathbf{l}$  are obtained, which are usually referred to as the equations of the  $\sigma$ -model. The application of such equations considerably simplifies the analysis of both linear and nonlinear dynamic effects in an antiferromagnet, see Refs.<sup>18,19</sup>. However, the advantage of these equations for describing macroscopic quantum effects is not so obvious. The Lagrangian obtained from the classical Landau-Lifshitz equations or from symmetry considerations cannot be used directly for describing the MQT effect taking into account the interference of instanton trajectories. It is probably for this reason that Golyshev and Popkov<sup>5</sup> used in their analysis of the CMQT effects a system of two equations for the sublattice magnetizations, whose analysis is much more complicated.

As a matter of fact, it is impossible in principle to reconstruct the Lagrangian of a dynamic system from the classical equations of motion. The Lagrangians describing the same classical equations of motion for the system can differ in the term which is the total derivative with respect to time. This term does not effect on the classical dynamics of the system, but alters the magnitude of the Euclidean action on trajectories. For this reason the corresponding terms with total derivatives were lost in the early publications.<sup>6,7</sup> A consistent quantum-mechanical expression for the spin Lagrangian taking into account the correct equation for the total derivative can be derived using the formalism of coherent states and the analysis of the evolution operator; this expression coincides with Eq. (1) given above. Topological terms of the form of total derivatives in the effective Lagrangian for the vector  $\mathbf{l}$  are significant for the quantum theory of 1D antiferromagnets.<sup>11</sup> However, it is impossible in principle to derive their expressions only from the classical equations of the  $\sigma$ -model for the vector  $\mathbf{l}$ .

In the simplest version of the  $\sigma$ -model the derivatives of  $\mathbf{l}$  with respect to time appear in the Lagrangian in the trivial form  $(\partial\mathbf{l}/\partial\tau)^2$ , see Refs.<sup>13–15</sup>. In this case the equations of the  $\sigma$ -model are Lorentz-invariant, and the description of the dynamics of nonlinear magnetization waves (kink-type solitons in antiferromagnets) is considerably simplified.<sup>13,19</sup> The interference effects in the MQT are obviously absent. However, the situation changes drastically for more realistic models.

First, for many antiferromagnetic crystals there exist terms reflecting interactions of the DM type, which

are linear in  $\mathbf{l}$  and in magnetization. It was shown in Refs.<sup>20,21</sup> that these interactions are responsible for the terms in the effective Lagrangian which are linear in  $\partial\mathbf{l}/\partial\tau$ ; this considerably modifies the kink dynamics in comparison with the simplest Lorentz-invariant model. Obviously, such interactions can also lead to the emergence of total derivatives (topological phases). The presence of a magnetic field may also lead to similar effects; this was noted in the analysis of the nonlinear dynamics of antiferromagnets<sup>22</sup> as well as for the MQT effect, see Ref.<sup>22</sup> and recent publications<sup>5,23–27</sup>.

In the present paper we will construct the Lagrangian of the  $\sigma$ -model on the base of the Eq. (1) taking into account all possible sources of the terms with the total derivative, which may lead to nontrivial interference effects. This Lagrangian will be used to study the interference of instanton trajectories for the real models of antiferromagnetic particles of various symmetries and to determine the contribution of these effects to the tunneling probability.

## II. LAGRANGIAN OF THE $\sigma$ -MODEL FOR REAL ANTIFERROMAGNETS

Let us consider a system with localized spins, in which nearest neighbors are coupled through the antiferromagnetic interaction. We assume that the lattice has such a structure that the sites with spins can be divided into two groups so that the spins appearing in pairs of nearest neighbors belong to different groups and there are no frustrations in the lattice. For ideal antiferromagnets these two groups correspond to the two magnetic sublattices. In this case we assume that the spins corresponding to each group have parallel orientations and form the total spins  $\mathbf{S}_1$  and  $\mathbf{S}_2$  of the sublattices. In the exchange approximation for such antiferromagnets, vectors  $\mathbf{S}_1$  and  $\mathbf{S}_2$  are antiparallel. The total spin  $\mathbf{S} = \mathbf{S}_1 + \mathbf{S}_2$  in the ground state can differ from zero in view of a different number of sites in the sublattices (decompensation),  $|\mathbf{S}_1| \neq |\mathbf{S}_2|$ , and also in the presence of an external magnetic field and/or the DM interaction, when the antiparallelism of the spins is violated (i.e.,  $|\mathbf{S}_1 + \mathbf{S}_2| \neq 0$  even for  $|\mathbf{S}_1| = |\mathbf{S}_2|$ ). We will consider only completely compensated antiferromagnets with  $|\mathbf{S}_1| = |\mathbf{S}_2|$ , since the specific effects associated with spin decompensation ( $|\mathbf{S}_1| \neq |\mathbf{S}_2|$ , but the difference  $|\mathbf{S}_1 - \mathbf{S}_2| \ll |\mathbf{S}_{1,2}|$ ) reduce the interference effects to those which are well known for ferromagnets.<sup>28</sup>

Our goal is to construct the Lagrangian describing the dynamics of the vector  $\mathbf{l}$  in the presence of the DM interaction and the magnetic field. Since the tensor of exchange interaction constants  $J_{ij}$  may have the antisymmetric component in the nearest neighbor approximation, the Hamiltonian of such a system has the form

$$\mathcal{H}_e = J \sum_{\langle\alpha\beta\rangle} \mathbf{S}_\alpha \mathbf{S}_\beta + \sum_{\langle\alpha\beta\rangle} \mathbf{d} \cdot (\mathbf{S}_\alpha \times \mathbf{S}_\beta) - g\mu_B \sum_{\alpha} \mathbf{H} \cdot \mathbf{S}_\alpha. \quad (2)$$

Here, the first term describes the isotropic exchange interaction, the summation in this term is extended to the pairs of nearest neighbors, and  $\mathbf{S}_\alpha$  is the spin at the  $\alpha$ th site. The antisymmetric component of the tensor of the exchange constants  $J_{ij}$  is a microscopic source of the DM interaction,<sup>29</sup> corresponding to the dual vector  $\mathbf{d}$ . The last term describes the interaction of spins with the external magnetic field.

Let us consider the exchange approximation in which the deviation from the conventional Heisenberg model with an isotropic exchange interaction  $J\mathbf{S}_\alpha\mathbf{S}_\beta$  is small; i.e.,  $d, g\mu_B H \ll J$ . In this case we can introduce the total spins  $\mathbf{S}_1 = \sum \mathbf{S}_{\alpha_1}$  and  $\mathbf{S}_2 = \sum \mathbf{S}_{\alpha_2}$  of the sublattices and assume that the vectors  $\mathbf{S}_1$  and  $\mathbf{S}_2$  have a fixed length. It is convenient to put  $\mathbf{S}_1 = Ns\boldsymbol{\sigma}_1$  and  $\mathbf{S}_2 = Ns\boldsymbol{\sigma}_2$ , where  $s$  is the spin at a site and  $N$  is the number of sites in each sublattice. We will parametrize the unit vectors  $\boldsymbol{\sigma}_1$  and  $\boldsymbol{\sigma}_2$  by the polar coordinates  $(\theta_1, \phi_1)$  and  $(\theta_2, \phi_2)$ , respectively. In this case the classical magnetic energy of the antiferromagnet, whose exchange component corresponds to the Hamiltonian (2), can be written in the form

$$\mathcal{W}(\boldsymbol{\sigma}_1, \boldsymbol{\sigma}_2) = Js^2 z N \boldsymbol{\sigma}_1 \boldsymbol{\sigma}_2 + s^2 z N \mathbf{d} \cdot (\boldsymbol{\sigma}_1 \times \boldsymbol{\sigma}_2) + w(\boldsymbol{\sigma}_1, \boldsymbol{\sigma}_2) - g\mu_B s N \mathbf{H}(\boldsymbol{\sigma}_1 + \boldsymbol{\sigma}_2). \quad (3)$$

Here,  $N$  is the number of spins in sublattices,  $z$  is the coordination number for a lattice site, and  $w(\boldsymbol{\sigma}_1, \boldsymbol{\sigma}_2)$  is the anisotropy energy.

Thus, we arrive at the description of the energy of an antiferromagnet in terms of two unit vectors. Their dynamics can be described by a Lagrangian which can be written in the dynamic variables  $\boldsymbol{\sigma}_1$  and  $\boldsymbol{\sigma}_2$  taking into account relation (3) in the form

$$\mathcal{L} = -i\hbar S_1 \mathbf{A}_1(\boldsymbol{\sigma}_1) \dot{\boldsymbol{\sigma}}_1 - i\hbar S_2 \mathbf{A}_2(\boldsymbol{\sigma}_2) \dot{\boldsymbol{\sigma}}_2 - \mathcal{W}(\boldsymbol{\sigma}_1, \boldsymbol{\sigma}_2). \quad (4)$$

Here, we have chosen a more general form of the kinetic terms as compared to relation (1). These terms can be presented through the vector potential of the field of a magnetic monopole:

$$\mathbf{A}_{1,2}(\boldsymbol{\sigma}) = \frac{\boldsymbol{\sigma} \times \mathbf{n}_{1,2}}{\sigma(\sigma + \boldsymbol{\sigma} \mathbf{n}_{1,2})}, \quad (5)$$

where  $\mathbf{n}_{1,2}$  are the quantization axes of coherent states for each sublattice. This potential has a singularity for  $\boldsymbol{\sigma} \mathbf{n} = -\sigma$ , i.e., on a certain half-line in the  $\boldsymbol{\sigma}$ -space. Usually, the “north pole” gauge with  $\mathbf{n} = \hat{\mathbf{e}}_z$  is used, in which the quantity  $\mathbf{A}(\boldsymbol{\sigma})\dot{\boldsymbol{\sigma}}$  assumes to have the familiar form (1). The potentials  $\mathbf{A}_{1,2}$  of the monopole field permit gauge transformations (such as a change in the position of spin quantization axes and, hence, singularities), which do not change the equations of motion, but make a contribution to the Lagrangian in the form of the total derivative of the function of spins  $\boldsymbol{\sigma}_1$  and  $\boldsymbol{\sigma}_2$  with respect to  $\tau$ , which can be significant for the description of interference effects. The kinetic terms for each sublattice can be written in individual gauges (in particular,

with different directions of the quantization axes  $\mathbf{n}_1$  and  $\mathbf{n}_2$ ).

We present the unit vectors  $\boldsymbol{\sigma}_1$  and  $\boldsymbol{\sigma}_2$  in terms of vectors  $\mathbf{l} = (\boldsymbol{\sigma}_1 - \boldsymbol{\sigma}_2)/2$  and  $\mathbf{m} = (\boldsymbol{\sigma}_1 + \boldsymbol{\sigma}_2)/2$  which are connected through the relations

$$\mathbf{l}^2 + \mathbf{m}^2 = 1 \quad \text{and} \quad \mathbf{m} \mathbf{l} = 0. \quad (6)$$

In the  $\sigma$ -model approximation, when the magnetic moment is small  $|\mathbf{m}| \ll |\mathbf{l}|$ , we can use simple transformations in order to present the Lagrangian (4) as a power expansion in  $\mathbf{m}$ . Confining the expansion to the terms linear in  $\mathbf{m}$ , we can write the kinetic terms in the form

$$\begin{aligned} & -i\hbar \mathbf{A}_1(\boldsymbol{\sigma}_1) \dot{\boldsymbol{\sigma}}_1 - i\hbar \mathbf{A}_2(\boldsymbol{\sigma}_2) \dot{\boldsymbol{\sigma}}_2 = \\ & = -i\hbar \dot{\mathbf{l}} [\mathbf{A}_1(\mathbf{l}) - \mathbf{A}_2(-\mathbf{l})] - i\hbar m_i \left[ \dot{\mathbf{l}} \frac{\partial \mathbf{A}_1(\mathbf{l})}{\partial l_i} + \dot{\mathbf{l}} \frac{\partial \mathbf{A}_2(-\mathbf{l})}{\partial l_i} \right] \\ & \quad - i\hbar \dot{\mathbf{m}} [\mathbf{A}_1(\mathbf{l}) + \mathbf{A}_2(-\mathbf{l})]. \end{aligned} \quad (7)$$

Here, the form of the vector-potentials  $\mathbf{A}_1$  and  $\mathbf{A}_2$  has not been specified yet. In particular, the quantization axes  $\mathbf{n}_1$  and  $\mathbf{n}_2$  have not been chosen. It is natural to choose the quantization axes  $\mathbf{n}_1$  and  $\mathbf{n}_2$  so that  $\mathbf{A}_1(\mathbf{l}) = \mathbf{A}_2(-\mathbf{l})$ , which is possible for  $\mathbf{n}_1 = -\mathbf{n}_2$ . In this case the singular term with  $d\mathbf{l}/d\tau$  vanishes, and the dynamic terms expansion starts with the term linear in  $\mathbf{m}$ , which can be written in the form

$$-i\hbar \dot{\mathbf{m}} \cdot (\mathbf{F} \times \dot{\mathbf{l}}) - i\hbar \frac{d}{d\tau} [\mathbf{m} \cdot \mathbf{A}_1(\mathbf{l})], \quad (8)$$

where

$$\mathbf{F}_i = \epsilon_{ijk} \left( \frac{\partial A_j}{\partial l_k} - \frac{\partial A_k}{\partial l_j} \right). \quad (9)$$

Thus, most of the arbitrariness in the choice of the gauge field  $\mathbf{A}$ , which takes place for ferromagnets, does not exist for antiferromagnets. The gauge-invariant quantity  $F_i$ , which has the meaning of a formal magnetic field associated with potential  $\mathbf{A}$ , is the magnetic monopole field  $\mathbf{F} = \mathbf{l}/|\mathbf{l}|^3$ . In the transition from Eq. (7) to the expression (8) the initial gauge arbitrariness turned out to be localized in the term with the total derivative  $d[\mathbf{m} \mathbf{A}(\mathbf{l})]/d\tau$ . Concerning this quantity, its contribution to the Euclidean action is obviously equal to zero in the case when an instanton trajectory misses the singular point of  $\mathbf{A}(\mathbf{l})$ . This condition can be easily satisfied if we choose the direction  $\mathbf{n} = \mathbf{n}_1 = -\mathbf{n}_2$  along the hard magnetization axis of the antiferromagnet. In this case the phase for a closed path on the unit sphere  $\mathbf{l}^2 = 1$ , which is formed by instanton trajectories, is independent of the position of the quantization axis  $\mathbf{n}$ .

Taking into account the condition  $\mathbf{m} \mathbf{l} = 0$ , we eliminate from the expression (4) the slave variable  $\mathbf{m}$ :

$$\mathbf{m} = \frac{\hbar}{2Js_z} \left[ \gamma (\mathbf{H}^{\text{eff}} - \mathbf{l}(\mathbf{H}^{\text{eff}} \cdot \mathbf{l})) - i\mathbf{l} \times \dot{\mathbf{l}} \right], \quad (10)$$

where  $\gamma = g\mu_B/\hbar$  is the gyromagnetic ratio and  $\mathbf{H}^{\text{eff}}$  is the effective field which is the sum of the external field

$\mathbf{H}$  and the DM field  $\mathbf{H}^{\text{DM}}$ . In the approximation chosen above, in which the DM interaction can be presented in the purely antisymmetric form  $\mathbf{d}(\boldsymbol{\sigma}_1 \times \boldsymbol{\sigma}_2) \propto \mathbf{d}(\mathbf{l} \times \mathbf{m})$ , the DM field can be written as  $\mathbf{H}^{\text{DM}} = z s(\mathbf{d} \times \mathbf{l})/(g\mu_B)$ .

The expression for  $\mathbf{m}$  is also valid for more general forms of the DM interaction, which cannot be reduced to a bilinear form in  $\boldsymbol{\sigma}_{1,2}$ . In particular, we will consider more general forms of the DM interaction of the type  $D_{ik}(\mathbf{l})m_i l_k$  which are observed for many crystals and are significant for the MQT effects. In this case the effective field in the expression for  $\mathbf{m}$  assumes the form  $H_i^{\text{DM}} = D_{ik}(\mathbf{l})l_k$ , and

$$H_i^{\text{eff}} = H_i^{(0)} + D_{ik}(\mathbf{l})l_k. \quad (11)$$

In this section we will not specify the form  $D_{ik}(\mathbf{l})$ . The approximation  $|\mathbf{m}| \ll |\mathbf{l}|$  used in the derivation of the  $\sigma$ -model is satisfied for  $\max(H, H^{\text{DM}}) \ll H_{ex}$ , where  $H_{ex} = Jsz/\mu_B$  is the exchange field. Substituting  $\mathbf{m}$  into the Lagrangian (4), we obtain the effective Lagrangian for the vector  $\mathbf{l}$  in the form

$$\begin{aligned} \mathcal{L} = & \frac{\hbar^2 N}{2Jz} \left[ \frac{1}{2} \dot{\mathbf{l}}^2 + i\gamma \mathbf{H}^{\text{eff}} \cdot (\mathbf{l} \times \dot{\mathbf{l}}) \right] - \mathcal{W}_a(\mathbf{l}) \\ & + \frac{2\mu_B^2 N}{Jz} \left\{ (\mathbf{H} \cdot \mathbf{l})^2 - \mathbf{H}^2 + 2\mathbf{H} \left[ \mathbf{l}(\mathbf{H}^{\text{DM}} \cdot \mathbf{l}) - \mathbf{H}^{\text{DM}} \right] \right\}. \end{aligned} \quad (12)$$

Here,  $\mathcal{W}_a(\mathbf{l})$  has the meaning of the effective anisotropy energy in which the additional term is  $(\mathbf{H}^{\text{DM}} \cdot \mathbf{l})^2 - (\mathbf{H}^{\text{DM}})^2$  taken into account along with the initial energy  $w(\mathbf{l}) = w(\boldsymbol{\sigma}_1, \boldsymbol{\sigma}_2)$  introduced above for  $\boldsymbol{\sigma}_1 = -\boldsymbol{\sigma}_2 = \mathbf{l}$ . The quantity  $\mathcal{W}_a(\mathbf{l})$  is obviously the real anisotropy energy determined from static measurements in weak fields, and there is no point in separating these contributions. We must simply use the expression for  $\mathcal{W}_a(\mathbf{l})$  which is determined by the crystal symmetry of the magnet. The specific form of the anisotropy energy for various antiferromagnets is given in the Table I. The terms in the braces describe the variation of the static energy of the antiferromagnet due to the external magnetic field. The first term, which is quadratic in the components of  $\mathbf{H}$ , is quadratic in  $\mathbf{l}$  and can also be presented as the field induced renormalization of the anisotropy energy. The second term, which is bilinear in the components of the external magnetic field  $\mathbf{H}$  and the DM field  $\mathbf{H}^{\text{DM}}$ , contains odd powers of the components of  $\mathbf{l}$  and describes the energy of the weak ferromagnetic moment induced by the DM interaction. (In particular, this term can be reduced to  $\mathbf{H} \cdot (\mathbf{d} \times \mathbf{l})$  for a purely antisymmetric DM interaction.) This term can completely remove the degeneracy of the classical ground state of the system, and the analysis of the MQT effects becomes meaningless. For this reason, it makes sense to take into account the external magnetic field and the DM interaction simultaneously only for certain selected orientations of the external field, when this term vanishes for vector  $\mathbf{l}$  directed along the easy magnetization axis of the antiferromagnet. Some of these ori-

entations of the field for orthorhombic antiferromagnets were considered in Ref.<sup>5</sup>.

Thus, we arrive at the following conclusions. The Lagrangian for the vector  $\mathbf{l}$  differs from the Lagrangian for the  $\sigma$ -model of ideal antiferromagnets<sup>11</sup> in the presence of a number of additional terms which play different roles in the description of CMQT. In contrast to the case of ferromagnets or antiferromagnets with different spins of the sublattices, the term with the total derivative can easily be eliminated. It is important to note that the external field and some forms of the DM interaction leads to the emergence of gyroscopic terms linear in  $d\mathbf{l}/d\tau$ . The emergence of these terms indicates the lowering of the actual dynamic symmetry of antiferromagnets in the presence of a magnetic field and/or the DM interaction.

The structure of the Lagrangian is such that the contribution of the DM interaction to the gyroscopic term can be taken into account by adding the DM field  $\mathbf{H}^{\text{DM}}$ , which is a function of  $\mathbf{l}$ , to the external magnetic field  $\mathbf{H}$ . Gyroscopic terms can make significant contributions to the probabilities of tunneling processes both by effecting on the structure of instanton solutions and by creating destructive interference of instanton trajectories. It will be proved below that, in contrast to the case of a ferromagnets or antiferromagnets with different spins of the sublattices, this interference is not of topological origin, but can also be given below. The examples of “pure” antiferromagnets in which tunneling can be completely suppressed due to the interference of instanton trajectories will be given below.

Deriving the Lagrangian (12), we neglected the possibility of inhomogeneous tunneling and, hence, the dependence of  $\mathbf{S}$  and  $\mathbf{l}$  on spatial coordinates was omitted from the very outset. The inclusion of such a dependence leads to the substitution  $N \rightarrow \int dV/a^3$ , where  $a$  is the lattice constant, and to the emergence of an additional term proportional to  $Ja^2(\nabla\mathbf{l})^2$  in the Euclidean action. A comparison of the inhomogeneity energy with the anisotropy energy leads to an estimate of the spatial inhomogeneity size on the order of  $\Delta_0 = a\sqrt{H_{ex}/H_{an}}$ , where  $H_{ex}$  and  $H_{an}$  are the exchange field and the anisotropy field and  $\Delta_0$  is the domain wall thickness. If the size of a particle is larger than  $\Delta_0$ , i.e.,  $N > N_c \simeq (\Delta_0/a)^3 \simeq (H_{ex}/H_{an})^{3/2}$ , we can assume a more advantageous inhomogeneous tunneling scenario, in which the level splitting weakly depends on  $N$  (or is even independent of it) for  $N > N_c$ . Although this question has not been discussed in the literature and its analysis is beyond the scope of the present publication, we will briefly consider it.

The value of  $N_c$  is too large for the tunneling effects to be observable for  $N > N_c$ . As a matter of fact,  $N$  in the tunneling exponent is multiplied by the susceptibility of the system, i.e., appears in the combination  $NH_{an}/H_{ex}$ , see Ref.<sup>10</sup>. The presence of this small parameter actually makes it possible to observe tunneling at ferritin particles with  $N \simeq 3.5 \cdot 10^3$ , see Refs.<sup>8,30</sup>. However, for typical values of  $H_{an}/H_{ex} \sim 10^{-2} - 10^{-3}$ , the tunneling exponent  $N_c H_{an}/H_{ex} \simeq (H_{ex}/H_{an})^{1/2} \gg 1$  is too large and

the observation of the transition to the inhomogeneous tunneling mode becomes problematic.

### III. SYMMETRY OF THE INSTANTON SOLUTIONS AND INTERFERENCE OF CONTRIBUTIONS FROM INSTANTON TRAJECTORIES

In accordance with the general rules of the semiclassical approximation formulated in the instanton language, the amplitude of transition from one state to another is described in the so-called instanton-gas approximation<sup>31</sup>. The level splitting for a system with two equivalent minima can be presented in the form

$$\Delta \propto D\sqrt{K}, \quad (13)$$

where the quantity  $D$  is defined as

$$D = (\det {}'\hat{\Omega})^{-1/2} \left( \frac{\text{Re} \mathcal{I}}{2\pi\hbar} \right)^{1/2} \exp\left(-\frac{\text{Re} \mathcal{I}}{\hbar}\right), \quad (14)$$

and  $\mathcal{I}$  is the one-instanton action;  $K$  is a combinatorial factor emerging due to nonuniqueness of the tunnel path connecting two equivalent minima; and  $\det {}'\hat{\Omega}$  is the fluctuation determinant disregarding the zeroth mode, which is determined by small deviations from an instanton trajectory, see Ref.<sup>31</sup> for details. In order to analyze the effects of tunneling between degenerate states corresponding to the ground states of the system and to determine the value of splitting, we must find the one-instanton trajectories connecting these states, calculate the value of the Euclidean action  $\mathcal{I}$  on these trajectories, and find the determinant of the operator for the second variation of action. The contribution to the splitting comes only from equivalent trajectories corresponding to the minimum value of the real component of  $\mathcal{I}$ . The combinatorial factor depending on the phase difference in the trajectories will be calculated below using Eq. (18). In this section, we concentrate our attention on an analysis of the main contribution which comes only from  $\mathcal{I}$  and will not calculate the preexponential factor. Let us see how these calculations can be carried out in actual practice.

For a concrete analysis, it is convenient to write the Lagrangian in the form

$$\mathcal{L} = \frac{\hbar^2 N}{2Jz} \left[ \frac{1}{2} \left( \frac{d\mathbf{l}}{d\tau} \right)^2 + i(\boldsymbol{\omega}_H \times \mathbf{l}) \cdot \frac{d\mathbf{l}}{d\tau} + \frac{\omega_0^2}{2} w_a(\mathbf{l}) \right], \quad (15)$$

where  $\boldsymbol{\omega}_H = \gamma \mathbf{H}^{\text{eff}}$ ,  $\gamma$  is the gyromagnetic ratio, and  $\mathbf{H}^{\text{eff}}$  is the effective field. The dimensionless function  $w_a(\mathbf{l})$  is proportional to the anisotropy energy, and the value of  $\omega_0$  coincides with the frequency of a homogeneous antiferromagnetic resonance in the uniaxial anisotropy field. We parametrize the vector  $\mathbf{l}$  by the angular variables

$$l_1 = \sin \theta \cos \phi, \quad l_2 = \sin \theta \sin \phi, \quad l_3 = \cos \theta. \quad (16)$$

We are dealing with an easy-axis anisotropy. Consequently, the ground state is doubly degenerated and has two values of  $\mathbf{l}$  corresponding to it:  $\mathbf{l} = \hat{\mathbf{e}}_3$  and  $\mathbf{l} = -\hat{\mathbf{e}}_3$ , and the unit vector  $\hat{\mathbf{e}}_3$  being parallel to the easy axis. Let us consider the tunneling between these two states. Function  $w_a(\mathbf{l})$  for a magnet with the anisotropy axis  $C_n$  can be written in the form

$$w_a(\theta, \phi) = \sin^2 \theta + \tilde{w}_a(\theta, \phi), \quad (17)$$

where the first term corresponds to easy-axis anisotropy and  $\tilde{w}_a(\theta, \phi) \ll 1$  defines anisotropy in the basal plane.

For antiferromagnets with an easy axis of symmetry  $C_n$ , there exist  $n$  instanton trajectories and  $n$  anti-instanton trajectories, and the combinatorial factor has the form

$$K = \sum_{k, \bar{k}'=0}^{n-1} \cos \Phi_{k, \bar{k}'}, \quad \Phi_{k, \bar{k}'} = \frac{1}{\hbar} \text{Im} \oint_{k \cup \bar{k}'} d\tau \mathcal{L}(\mathbf{l}, \dot{\mathbf{l}}), \quad (18)$$

i.e.,  $\Phi_{k, \bar{k}'}$  is the phase difference between the  $k$ th instanton and the  $k'$ th anti-instanton. The integral defining  $\Phi_{k, \bar{k}'}$  is taken over a closed path formed by the trajectories of the  $k$ th instanton and the  $\bar{k}'$ th anti-instanton. In the Lorentz-invariant  $\sigma$ -model, the Lagrangian is real and all  $\Phi_{k, \bar{k}'}$  are equal to zero; the combinatorial factor  $K = n^2$  is trivial and equal to  $n$ . Consequently,  $\sqrt{K} = n$ ; i.e., the total transition amplitude and level splitting for  $nn$  pairs is just the contribution from one instanton multiplied by the number of paths. It will be shown below, however, that for  $\Phi_{k, \bar{k}'} \neq 0$ , the level splitting  $\Delta$  may contain an oscillatory dependence on the product of the small parameter  $|\boldsymbol{\omega}_H|$  and the large quantity  $N$ , and, hence, requires a more detailed analysis. The nature of its oscillations can be established from symmetry considerations, and the specific form of the function  $K$  of the parameters of the problem can be determined even without solving the corresponding the Euler-Lagrange equations.

#### A. Lorentz-invariant $\sigma$ -model

It is convenient to consider first the tunneling in the simplest Lorentz-invariant  $\sigma$ -model which corresponds to the Lagrangian (15) with  $\boldsymbol{\omega}_H = 0$ . As a matter of fact, for some models of an antiferromagnet with the DM interaction, the results turn out to be the same as in the absence of this interaction, see below. If  $H^{\text{eff}} = 0$ , the analysis of the problem does not present any difficulty. Indeed, for any form of the anisotropy energy in a uniaxial antiferromagnet with the principal axis  $C_2, C_4, C_6$  (in the subsequent analysis, we will consider only the type of symmetry that can exist in the crystal lattice), the instanton solution corresponds to the function  $\theta = \theta(\tau)$  with the boundary conditions  $\theta \rightarrow 0, \pi$  for  $\tau \rightarrow \pm\infty$  and

$\phi = \phi_0 = \text{const}$ , where  $\phi_0$  is defined by the relation

$$\left. \frac{\partial w_a(\theta, \phi)}{\partial \phi} \right|_{\phi=\phi_0} = 0. \quad (19)$$

Let us assume that the ground states  $\pm \hat{e}_3$  are on the principal axis  $C_n$ . In this case the value of  $\widetilde{w}_a(\theta, \phi)$  is proportional to  $\sin n\phi$  and there exist  $2n$  solutions to this equation:

$$\phi_k^{(0)} = \frac{\pi k}{n}, \quad k = 0, 1, \dots, 2n-1, \quad (20)$$

from which  $n$  solutions  $\phi_{k,\min}^{(0)}$  correspond to the minima of  $w_a(\theta, \phi)$ , while the remaining  $n$  solutions  $\phi_{k,\max}^{(0)}$  correspond to the maxima of this function for all  $\theta \neq 0, \pi$ . Instantons with  $\phi_{k,\min}^{(0)}$  correspond to the lowest value of the Euclidean action, and we will consider below only these  $n$  solutions. Function  $\theta(\tau)$  can be determined from the second-order equation for which the first integral is known to be

$$\left( \frac{d\theta}{d\tau} \right)^2 = \omega_0^2 [w_a(\theta, \phi_k^{(0)}) - w_a(0, \phi_k^{(0)})]. \quad (21)$$

Henceforth, we assume that  $w_a(0, \phi) = w_a(\pi, \phi) = 0$  and that the value of  $\phi = 0$  corresponds to the minimum of the function  $w_a(\theta, \phi)$ . With such a choice of the axes,  $z$  is always an easy magnetization axis and  $x$  is an medium magnetization axis. The Euclidean action on trajectories is real-valued for all values of  $\phi$  and is defined as

$$\mathcal{I} = \frac{\hbar^2 \omega_0 N}{2Jz} \int_0^\pi d\theta \sqrt{w_a(\theta, \phi)}. \quad (22)$$

This approximate expression is written in the main approximation in small anisotropy in the basal plane  $\tilde{\beta} \ll 1$ , where  $\tilde{\beta}$  is the characteristic anisotropy constant in the basal plane, i.e., the maximum value of  $\widetilde{w}_a$ . Thus, the contribution in the given case comes from  $n$  instanton trajectories on which vector  $\mathbf{l}$  is real and rotates in one of the  $n$  planes defined by the condition  $\phi = \phi_{k,\min}^{(0)} \equiv \phi_k^{(0)}$ . The imaginary component of  $\mathcal{I}$  in the Lorentz-invariant model is absent, and the combinatorial factor  $K$  in the expression (18) is equal to  $n^2$ .

### B. Role of $H^{\text{eff}}$

The inclusion of the terms with  $H^{\text{eff}}$ , which destroy the Lorentz invariance, brings about two types of difficulties. First, for  $H^{\text{eff}} \neq 0$ , the solution  $\phi = \text{const}$  is generally inapplicable, and the instanton structure is determined by the general system of two second-order equations

$$-\ddot{\theta} + \dot{\phi}^2 \sin \theta \cos \theta + \omega_0^2 \frac{\partial w_a}{\partial \theta} + i\omega_H \dot{\phi} \Gamma(\theta, \phi) = 0, \quad (23a)$$

$$-\ddot{\phi} \sin^2 \theta - 2\dot{\phi}\dot{\theta} \sin \theta \cos \theta + \omega_0^2 \frac{\partial w_a}{\partial \phi} - i\omega_H \dot{\theta} \Gamma(\theta, \phi) = 0, \quad (23b)$$

whose solutions are generally not real-valued. Here, the terms with  $\Gamma$  are determined by the variation of the term with  $H^{\text{eff}} \cdot [(d\mathbf{l}/d\tau) \times \mathbf{l}]$  in the Lagrangian (15), and the form of the function  $\Gamma(\theta, \phi)$  generated by the DM interaction for various types of magnetic symmetry is given in the column 5 of the Table I. Second, the imaginary component of the Euclidean action  $\mathcal{I}$ , which comes from the term proportional to  $\omega_H$ , may appear even for trajectories with a real  $\mathbf{l}$ . Let us consider the cases when these situations are realized.

If  $\Gamma(\theta, \phi)$  vanishes at the same values of  $\phi_k^{(0)}$  as for  $\partial w_a(\theta, \phi)/\partial \phi$ , the second equation (23b) is satisfied identically for the plane trajectories  $\dot{\phi} = 0$ , while the first equation in the system (23a) can be reduced to Eq. (21) considered above in the Lorentz-invariant  $\sigma$ -model. Consequently, in this case  $\Gamma(\theta, \phi)$  does not effect on the form of the function  $\theta = \theta(\tau)$  in the instanton solution, but changes the imaginary component of the Euclidean action. This effect will be considered in more details in the section IV.

If, however,  $\Gamma(\theta, \phi_k^{(0)}) \neq 0$ , the instanton does not correspond to a plane solution  $\phi = \text{const}$  any longer, and we must seek the general solution of the system (23) in the form  $\theta = \theta(\tau)$ ,  $\phi = \phi(\tau)$ . In this case the functions  $\theta(\tau)$  and  $\phi(\tau)$  may in general turn out to be complex-valued. There are no general analytical methods for constructing such separatrix solutions; an instanton solution of the system of equations (23) can be written exactly only for some cases (see Ref.<sup>32</sup> and the section IV in the present paper).

It will be shown below that the effect of the term in the Lagrangian on the imaginary component of the Euclidean action  $\mathcal{I}$  may lead to nontrivial consequences even for antiferromagnets with  $\Gamma(\theta, \phi) \neq 0$ , but  $\Gamma(\theta, \phi_k^{(0)}) = 0$ , and there exists a real-valued instanton solution  $\theta = \theta(\tau)$ ,  $\phi = \phi_k^{(0)}$ , or in the case when the value of  $H^{\text{eff}}/H_{ex}$  is negligibly small or its inclusion changes  $\theta = \theta(\tau)$  and the real component of  $\mathcal{I}$  insignificantly.

In order to explain this fact, we consider the case when the value of  $\Gamma/H_{ex} \ll 1$  is so small that instanton trajectories can be regarded as planar,  $\theta = \theta(\tau)$ ,  $\phi = \text{const}$ . The presence of the term linear in  $d\mathbf{l}/d\tau$  leads to the contribution to the imaginary component of the Euclidean action  $\mathcal{I}$ , which is proportional to the number of spins in the particle. The imaginary component of the Euclidean action  $\mathcal{I}$  is of order of  $\text{Im} \mathcal{I}/\hbar \propto Nd/J$ ; i.e., it is proportional to the product of a small and a large parameter. Consequently, the effects of destructive interference can be significant. It is well known that the interference effects for orthorhombic ferromagnets may suppress tunneling completely.<sup>1-4</sup> In contrast to the case of antiferromagnets the term with  $d\mathbf{m}/d\tau$  for ferromagnets does not contain a small factor  $H^{\text{eff}}/H_{ex}$ , but it is inessential since the value of  $\text{Im} \mathcal{I}/\hbar \simeq \pi Ns \gg 1$  for ferromagnets, while tunneling is completely suppressed when  $\text{Im} \mathcal{I}/\hbar \simeq \pi$ . This condition can easily be satisfied for a large  $N$ . In particular, for the antiferromagnetic particle of the ferritin with  $N \simeq 3500$  the tunneling probability

TABLE I: Anisotropy in the basal plane and the Dzyaloshinskii-Moriya interaction constant for systems with various types of the magnetic symmetry.

1	2	3	4 <sup>a</sup>	5	6 <sup>b</sup>	7	8
$n$	$\tilde{w}_a$	Axes	DMI	$\Gamma(\theta, \phi)$		$B(\theta, \phi)$	$K$
2	$\beta_2 \sin^2 \theta \sin^2 \phi$	$2_z^{(+)} 2_x^{(-)} 2_y^{(-)}$	$m_x l_y + m_y l_x$	$3 \sin^3 \theta \sin 2\phi$	*	$3 \sin^2 \theta \sin 2\phi$	4
		$2_z^{(-)} 2_x^{(+)} 2_y^{(-)}$	$m_y l_z + m_z l_y$	$6 \sin^2 \theta \cos \theta \sin \phi$	*	$6 \sin \theta \cos \theta \sin \phi$	4
		$2_z^{(-)} 2_x^{(-)} 2_y^{(+)}$	$m_x l_z + m_z l_x$	$6 \sin^2 \theta \cos \theta \cos \phi$		$6 \sin \theta \cos \theta \cos \phi$	4
4	$\beta_4 \sin^4 \theta \sin^2 2\phi$	$4_z^{(+)} 2_x^{(-)} 2_{xy}^{(-)}$	$\frac{1}{2i}(m_+ l_+^3 - m_- l_-^3)$	$5 \sin^5 \theta \sin 4\phi$	*	$5 \sin^4 \theta \sin 4\phi$	16
		$4_z^{(-)} 2_x^{(+)} 2_{xy}^{(-)}$	$m_x l_x - m_y l_y$	$3 \sin^3 \theta \cos 2\phi$		$3 \sin^2 \theta \cos 2\phi$	16
		$4_z^{(-)} 2_x^{(-)} 2_{xy}^{(+)}$	$m_x l_y + m_y l_x$	$3 \sin^3 \theta \sin 2\phi$	*	$3 \sin^2 \theta \sin 2\phi$	$8 + 8 \cos(sdN/J)$
6	$\beta_6 \sin^6 \theta \sin^2 3\phi$	$6_z^{(+)} 2_x^{(-)} 2_{\pi/6}^{(-)}$	$\frac{1}{2i}(m_+ l_+^5 - m_- l_-^5)$	$7 \sin^7 \theta \sin 6\phi$	*	$7 \sin^6 \theta \sin 6\phi$	36
		$6_z^{(-)} 2_x^{(+)} 2_{\pi/6}^{(-)}$	$\frac{1}{2i} m_z (l_+^3 - l_-^3)$	$5 \sin^4 \theta \cos \theta \sin 3\phi$	*	$5 \sin^3 \theta \cos \theta \sin 3\phi$	36
		$6_z^{(-)} 2_x^{(-)} 2_{\pi/6}^{(+)}$	$\frac{1}{2} m_z (l_+^3 + l_-^3)$	$5 \sin^4 \theta \cos \theta \cos 3\phi$		$5 \sin^3 \theta \cos \theta \cos 3\phi$	36

<sup>a</sup>For high-order axes, the following notation is introduced:  $m_{\pm} = m_x \pm i m_y$  and  $l_{\pm} = l_x \pm i l_y$ .

<sup>b</sup>Asterisks mark systems for which an exact solution corresponding to the minimum of the real part of the action exist.

in the magnetic field with interference is an oscillating function of the field, and the suppression of tunneling can be observed in fields  $H \lesssim 100$  Oe, see Refs.<sup>5,28,33</sup>, which are much weaker than the characteristic value of the DM field  $H^{\text{DM}} = 10^3 - 10^5$  Oe.

On the other hand, the contribution to the real component of the Euclidean action does not contain the large parameter  $N$ . This contribution can be appreciable, see the next section, but in this case the product of other parameters, namely, the small quantity  $d/J \ll 1$  and the large quantity  $d/\tilde{\beta} \gg 1$ , is significant. Thus, the terms with  $d\mathbf{l}/d\tau$  may lead to two types of effects: (i) the emergence of nonplanar instanton trajectories and complex values of components of  $\mathbf{l}$  on these trajectories; (ii) the interference of instantons even in the case of plane trajectories with the real Euclidean action  $\mathcal{I}$ .

The first effect only takes place when the term  $\Gamma(\theta, \phi)$  in Eq. (23) differs from zero. Such terms are always important for the description of the domain wall dynamics in antiferromagnets: they may reduce the limit velocity of the domain wall motion to a considerable extent and may also lead to an abrupt change in the wall structure upon a continuous variation of its velocity.<sup>20,21</sup> The subsequent analysis of concrete instanton solutions will show that the role of such terms in the description of the instanton structure and tunneling is not so important as in the description of the domain wall dynamics. On the other hand, if the function  $\Gamma(\theta, \phi)$  differs from zero, but the function  $\phi = \phi_k^{(0)}$  for the given solution  $\Gamma(\theta, \phi_k^{(0)}) = 0$ , the domain wall dynamics is trivial and can be described by Lorentz-invariant expressions. In

this case, the instanton structure  $\theta = \theta(\tau)$  is the same as in the Lorentz-invariant theory. However, the situation with instantons is different: not all features can be described by the function  $\theta(\tau)$  and the real component of the Euclidean action  $\mathcal{I}$  only. It will be shown below that the main contribution from the term  $\mathbf{H}^{\text{eff}} \cdot [(d\mathbf{l}/d\tau) \times \mathbf{l}]$  is associated precisely with interference processes and is manifested most clearly exactly when the instanton trajectory is planar; i.e.,  $\Gamma(\theta, \phi^{(0)}) = 0$ .

In the case of real-valued trajectories, it is convenient to use the following approach for calculating the imaginary component of the Euclidean action.<sup>34</sup> We introduce the vector  $\mathbf{r} = r\mathbf{l}$  which is not subjected to the condition  $r^2 = 1$  and present the term with the first derivative in the expression (15) in the form

$$-i\gamma \mathcal{A} \frac{\partial \mathbf{r}}{\partial \tau}, \quad \text{with} \quad \mathcal{A} = \frac{\mathbf{r} \times \mathbf{H}^{\text{eff}}}{r^2}. \quad (24)$$

This expression has the same structure as the term in the nonrelativistic Lagrangian describing the interaction of a classical charged particle moving in a 3D space with coordinate  $\mathbf{r}$  and velocity  $\mathbf{v} = d\mathbf{r}/d\tau$  with a formal magnetic field  $\mathbf{B} = \nabla \times \mathcal{A}$  (differentiation is carried out in the  $\mathbf{r}$  space). It is well known that the magnetic field appears in the Lagrangian of a charged particle through the vector potential  $\mathcal{A}$  at point  $\mathbf{r}$ , which is defined only to within a certain gauge, while the field  $\mathbf{B}$  is gauge-invariant.

Simple but cumbersome calculations proved that, for any ferromagnet, this formal magnetic field  $\mathbf{B}$  may be

radial and can be presented in the form

$$\mathbf{B} = \frac{\mathbf{r}}{r^2} B(\theta, \phi), \quad (25)$$

where

$$B(\theta, \phi) = 2(\mathbf{H}^{\text{eff}} \mathbf{l}) - \frac{\partial H_i^{\text{eff}}}{\partial l_i} + \frac{\partial H_i^{\text{eff}}}{\partial l_k} l_i l_k. \quad (26)$$

In the absence of the DM interaction the value of  $B(\theta, \phi)$  is determined only by the external field  $\mathbf{H}^{(0)}$ ,  $B(\theta, \phi) = 2(\mathbf{H}^{(0)} \mathbf{l})$ . At zero field the value of  $B(\theta, \phi)$  is determined by the DM field  $H_i^{\text{DM}} = D_{ij}(\mathbf{l}) l_j$  and can be presented in terms of the tensor  $D_{ij}$  as

$$B(\theta, \phi) = 3D_{ij} l_i l_j - D_{ii} + D_{ij,k} l_i l_j l_k - D_{ij,i} l_j. \quad (27)$$

Here, the comma in the subscript in  $D$  indicates the differentiation of the tensor  $D_{ij}$  with respect to the corresponding component of  $\mathbf{l}$ , and the summation is extended over double indices. The values of  $B(\theta, \phi)$  for various types of DM interaction and of the configurations of axes are given in column 7 of the Table I.

Phases  $\Phi_{k,\bar{k}'}$  defined in Eq. (18) can be presented in terms of the integrals  $\int \mathbf{A} d\mathbf{r}$  taken over instanton-antiinstanton pairs forming closed loops. Using the Stokes theorem, we can present the phase difference  $\Phi_{k,k'}$  as the magnetic flux of the vector  $\mathbf{B}$  through a part of the unit sphere bounded by such a loop. Obviously, individual phases are determined by the vector potential  $\mathbf{A}$ , i.e., depend on the gauge, but the phase differences are gauge-invariant for all pairs of trajectories.

It is important that the structure of  $B(\theta, \phi)$  for all possible types of DM interaction is such that the total flux of field  $\mathbf{B}$  through the unit sphere,

$$\Phi_{\text{tot}} = \int_0^\pi \sin \theta d\theta \int_0^{2\pi} d\phi B(\theta, \phi) \quad (28)$$

is equal to zero and the value of  $\cos \Phi_{k,\bar{k}'}$  is independent of the total time derivative in the Lagrangian (gauge invariance).<sup>39</sup>

This approach enables us to calculate specifically the phase difference of integral trajectories and the combinatorial factor  $K$  in Eq. (13). We begin with the simplest case of an orthorhombic antiferromagnet for which there are only two pairs of instanton trajectories. It can be easily verified that the DM interaction of any type (see the Table I) makes a zero contribution to the phase difference for two diametrically opposite trajectories.<sup>34</sup> For this reason the required phase factor can be determined only by the external field and can be written in the form

$$\cos \Phi = \cos \left( \frac{g \mu_B H^{(0)} N s}{J z} \cos \alpha \right), \quad (29)$$

where  $\alpha$  is the angle between the plane containing the instanton trajectories and the external field  $\mathbf{H}^{(0)}$ . This result was obtained by Chiolero and Loss<sup>33</sup> in particular

cases  $\alpha = 0$  and  $\alpha = \pi/2$ . Thus, for instanton trajectories lying in the same plane all possible types of DM interaction given in the Table I do not effect on the tunneling. This results does not contradict the analysis carried out by Golyshev and Popkov<sup>5</sup>, who studied tunneling in small completely compensated particles of an orthorhombic antiferromagnet with the orthoferrite structure and discovered that no interference effects exist in a zero magnetic field. The our approach enabled us to obtain this result without solution of the Euler-Lagrange equations. Thus, we have proved that the conclusion that no interference takes place for diametrically opposite trajectories can be extended to more general cases of the DM interaction. It is important that this conclusion is drawn without resorting to any approximation, which is inevitable in the solution of a complex system of equations describing the instanton structure.

Thus, in the case of orthorhombic antiferromagnets with two instanton trajectories none of the types of DM interaction presented in the Table I leads to destructive interference. However, this result is different for uniaxial antiferromagnets with an easy magnetization axis  $C_n$ ,  $n > 2$ . In this case there exist  $n$  pairs of instanton trajectories. Obviously, here we also have pairs of trajectories lying in the same plane, for which  $\Phi_{k,\bar{k}'} = 0$  and interference is trivial, but it can be manifested itself for pairs of trajectories with  $\phi_k^{(0)} - \phi_{k'}^{(0)} \neq \pi$ .

It will be shown below that the value of combinatorial factor can be reduced considerably from its maximum value  $n^2$  to zero; i.e., both partial and complete suppression of interference are possible.

### C. Tetragonal antiferromagnets

Let us demonstrate it using specific examples of particles with a tetragonal easy magnetization axis and binary axes in the perpendicular plane (crystallographic class  $4_z 2_x 2_{xy}$ ), when the minimum of the real component of the Euclidean action corresponds to four instanton and four antiinstanton trajectories. In order to describe tetragonal antiferromagnets, we choose the polar axis  $\hat{e}_3$  along the tetragonal easy magnetization axis  $4_z$ . Anisotropy in the basal plane is determined by the fourth-order invariant, see the Table I. We assume that  $\beta_4 > 0$ ; i.e., instanton trajectories correspond to the rotation of  $\mathbf{l}$  in the equivalent planes  $zx$  and  $zy$ . Depending on the magnetic parity of the principal axis (according to Turov<sup>36</sup>) and of the binary axes  $2_x$ ,  $2_y$ , or  $2_{xy}$ ,  $2_{yx}$  basically different types of behavior can be observed. We will consider them separately.

#### 1. System $4_z^{(+)} 2_x^{(-)} 2_{xy}^{(-)}$

With such a structure of axes in an antiferromagnet only the antisymmetric invariant  $d(m_x l_y - m_y l_x)$  is usually considered, which can be obtained from the antisym-



metric component of the tensor of exchange constants. The value of  $d$  is of order of  $\sqrt{\beta J}$ , see Ref.<sup>29</sup>. This invariant determines the weak isotropic ferromagnetic moment when  $\mathbf{l}$  is oriented in the basal plane. However, it is of no interest to us since it can be reduced to the total derivative in the Lagrangian and, hence, gives  $\Gamma(\theta, \phi) = 0$  in the equations of motion and  $B(\theta, \phi) = 0$  in the imaginary part of the Euclidean action. In addition, there exist a number of invariants of the relativistic origin,<sup>37</sup> which give a nonzero value of  $\Gamma(\theta, \phi)$ . The simplest of these invariants has the form  $2(l_x^2 - l_y^2)(m_x l_y + m_y l_x)$  which coincides (except for the total derivative) with the invariant  $(m_+ l_+^3 - m_- l_-^3)/(2i)$  presented in the table. It can easily be seen, however, that in this case  $\Gamma(\theta, \phi_k^{(0)}) = 0$ , and the instanton solution has the form  $\theta = \theta(\tau)$ ,  $\phi = \phi_k^{(0)} = \pi k/2$  for integer  $k$ . The value of  $B(\theta, \phi)$  is such that

$$B \propto \sin^4 \theta \sin 4\phi, \quad (30)$$

and, hence, all phases  $\Phi_{k, \bar{k}'}$  are equal to zero. An analysis of other invariants, e.g.,  $l_z^2(m_x l_y - m_y l_x)$ , leads to the same result (namely, the DM interaction does not effect on tunneling in any way). This result is apparently independent of the model and is determined only by the type of magnetic symmetry. The model independence for dynamic effects in domain walls has the same origin, i.e., the DM interaction; it was demonstrated in Refs.<sup>20,21</sup>. Thus, the case of an even principal axis  $4_z^{(+)}$  may serve as an example that nonzero terms which are linear in  $d\mathbf{l}/d\tau$  and cannot be reduced to the total derivative are not appeared in the separatrix solution and do not effect on tunneling in any way.

A different situation takes place for antiferromagnets with an odd principal axis  $4_z^{(-)}$ . In this case two cases are possible: when the intermediate anisotropy axes through which tunneling takes place are odd and when these axes are even.

### 2. System $4_z^{(-)} 2_x^{(-)} 2_{xy}^{(+)}$

In this case  $\Gamma(\theta, \phi) = 0$  and  $\phi = \phi_k^{(0)} = \pi k/2$ . The presence of the DM interaction does not effect on the instanton trajectories with the minimum action, which correspond to  $\phi = \phi_k^{(0)}$ ,  $\theta = \theta(\tau)$ . However, in contrast to the system with  $4_z^{(+)}$ , the contribution of the DM interaction is significant for calculating the combinatorial factor  $K$  in Eq. (13). It can be seen from the explicit expression  $B(\theta, \phi) \propto \sin^2 \theta \sin 2\phi$  that the phase difference for adjacent trajectories (with  $\phi = \phi_k^{(0)}$ ,  $\phi = \phi_{k\pm 1}^{(0)}$ ) differs from zero.

Thus, the phase factor for the tunneling probability is given by

$$K = 16 \sin^2 \left( \frac{sdN}{J} \right). \quad (31)$$

It is an oscillating function of the DM interaction constant  $d$ , and it takes the values in the range from 0 to 16. For realistic values of  $N$  of order of  $10^3 - 10^5$  the period is not large; the value of  $\Delta H^{\text{DM}}/H^{\text{DM}} \simeq 10^{-3} \div 10^{-1}$  for characteristic values of  $H^{\text{DM}} \simeq 10^4$  Oe and  $H_{ex} \simeq 10^6$  Oe. Since the value of the DM field is very sensitive to extrinsic parameters (e.g., the pressure or the addition of a small amount of impurities to the crystal), these oscillations can be observed and monitored. An additional opportunity for observing interference effects appears when the magnetic field is taken into consideration.

### 3. System $4_z^{(-)} 2_x^{(+)} 2_{xy}^{(-)}$

Such a symmetry group is typical for the extensively studied weak antiferromagnet  $\text{MnF}_2$ , see Ref.<sup>38</sup>. In this case  $\Gamma(\theta, \phi) \propto \sin^3 \theta \cos 2\phi$  and  $\Gamma(\theta, \phi) \neq 0$  for all values of  $\phi = \phi_k^{(0)}$ , corresponding to the minimum of the anisotropy in the basal plane and describing instanton trajectories for  $d = 0$ . For  $d \neq 0$  instanton solutions cannot be written in the simple form  $\theta = \theta(\tau)$ ,  $\phi = \pi k/2$ ,  $k = 0, 1, 2, 3$ . On the other hand, if we assume that the value of  $d$  is very small, we can easily find, applying the approximation of planar rotation and using the formula  $B(\theta, \phi) \propto \sin^2 \theta \cos 2\phi$ , that the difference in the imaginary components of  $\mathcal{I}$  for pairs of trajectories lying in the same plane as well as for adjacent instanton trajectories is equal to zero, and no interference effects take place. We will consider the solution for this case in the next section and prove that these simple regularities are preserved in a more rigorous analysis also when we do not require that  $\phi = \pi k/2$ . We will also consider a general mechanisms of the tunneling in the case when  $\Gamma(\theta, \phi_{\min}^{(0)}) \neq 0$  and the instanton solution is not real-valued.

## D. Hexagonal antiferromagnets

Let us briefly consider the case of a hexagonal principal axis. Here, we again have three cases presented in the Table I. For a system with an even principal axis  $6_z^{(+)} 2_x^{(-)} 2_{\pi/6}^{(-)}$  there exists the invariant  $m_x l_y - m_y l_x$  and the DM interaction, which cannot be reduced to a total derivative, appears only in the fifth order in  $\mathbf{l}$ . The analysis is similar to that for the system  $4_z^{(+)} 2_x^{(-)} 2_{xy}^{(-)}$ . In this case also nonzero terms which are linear in  $d\mathbf{l}/d\tau$  and cannot be reduced to a total derivative do not effect on tunneling in any way. It can be verified that such a behavior is the same as for antiferromagnets with the even principal axis  $n_z^{(+)}$ .

For systems with an odd principal axis, the DM interaction is a cubic function of  $\mathbf{l}$ , but it makes a zero contribution to the imaginary component of the Euclidean action under the assumption that instanton trajectories are real and planar. In the system  $6_z^{(-)} 2_x^{(-)} 2_{\pi/6}^{(+)}$  minimal

instanton trajectories have an imaginary component, but this only changes the real part of the Euclidean action. The analysis of this system is similar to that which will be carried out in the section IV for the system  $4_z^{(-)}2_x^{(+)}2_{xy}^{(-)}$ . Consequently, the combinatorial factor  $K$  for all three cases has the maximum value equal to  $K = 36$ .

#### IV. INSTANTON SOLUTION FOR ANTIFERROMAGNETS WITH THE SYMMETRY $4_z^{(-)}2_x^{(+)}2_{xy}^{(-)}$

It was noted above that in the case of antiferromagnets with the symmetry  $4_z^{(-)}2_x^{(+)}2_{xy}^{(-)}$  there is no exact solution of the type  $\phi = \pi k/2$ ,  $\theta = \theta(\tau)$  for trajectories with the rotation of  $\mathbf{l}$  in the vicinity of the easy plane  $\phi \simeq \pi k/2$ , and we have to analyze the complete system of two equations (23a) and (23b). The situation is complicated further since these equations have complex-valued coefficients and, in general, their complex solutions of the type  $\theta = \theta_1(\tau) + i\theta_2(\tau)$ ,  $\phi = \phi_1(\tau) + i\phi_2(\tau)$  must be considered. As a result, system (23) is equivalent to a dynamic system with four degrees of freedom, and it is not integrable. No general method exists for an analysis of such systems. However, a comprehensive analysis can be carried out in the given case as well as for magnets with other types of symmetry, which are listed in the Table I.

In the case of antiferromagnets with the odd tetragonal axis, the equations for the angular variables  $\theta$  and  $\phi$  have the form

$$-\ddot{\theta} - \dot{\phi}^2 \sin \theta \cos \theta + \omega_0^2 \sin \theta \cos \theta (1 + \beta_4 \sin^2 \theta \sin^2 2\phi) + 3i\omega_D \dot{\phi} \sin^3 \theta \cos 2\phi = 0, \quad (32a)$$

$$-\ddot{\phi} \sin^2 \theta - 2\dot{\phi}\dot{\theta} \sin \theta \cos \theta + \beta_4 \omega_0^2 \sin^4 \theta \sin 2\phi \cos 2\phi - 3i\omega_D \dot{\theta} \sin^3 \theta \cos 2\phi = 0. \quad (32b)$$

The quantity  $\omega_0$  defines the height of the potential barrier through which tunneling occurs, and  $\beta_4$  is the dimensionless anisotropy constant in the basal plane. We assume that  $\beta_4 > 0$ , which corresponds to instantons of the Lorentz-invariant  $\sigma$ -model ( $\omega_D = 0$ ) that passing through the even axes  $x$  or  $y$ ;  $\beta_4 \ll 1$  corresponds to the easy-axis limit, and  $\omega_D$  is proportional to the DM interaction constant,  $\omega_D = \gamma|\mathbf{H}^{\text{DM}}| = zd/\hbar$ .

It can be easily seen that this system has the exact solution  $\phi = \pi(2k+1)/4$ ,  $\theta = \theta(\tau)$ , which has been considered in the previous section. It determines tunneling for  $\beta_4 < 0$ , but in the case of  $\beta_4 > 0$  we are interested in now, it corresponds to the rotation of  $\mathbf{l}$  in the hard planes, does not ensure that the real part of the Euclidean action has a minimum, and makes a zero contribution to the tunneling amplitude in the instanton approximation. The exact solution  $\phi = \pi k/2$  does not exist in this case. It can be seen, however, that the substitution  $\phi = \pi k/2 + if(\tau)$  and  $\theta = \theta(\tau)$  with the real

functions  $f(\tau)$ ,  $\theta(\tau)$  and does not contradict this system and leads to the following system of two equations for functions  $f$  and  $\theta$ :

$$-\ddot{\theta} - \dot{f}^2 \sin \theta \cos \theta + \omega_0^2 \sin \theta \cos \theta (1 - \beta_4 \sin^2 \theta \sinh^2 2f) - 3(-1)^k \omega_D \dot{f} \sin^3 \theta \cosh 2f = 0, \quad (33a)$$

$$\ddot{f} \sin^2 \theta + 2\dot{f}\dot{\theta} \sin \theta \cos \theta - \beta_4 \omega_0^2 \sin^4 \theta \sinh 2f \cosh 2f + 3(-1)^k \omega_D \dot{\theta} \sin^3 \theta \cosh 2f = 0. \quad (33b)$$

In addition, such a substitution renders the Lagrangian real-valued:

$$\mathcal{L} = \frac{\hbar^2 N}{2Jz} \left[ \frac{\dot{\theta}^2 - \dot{f}^2 \sin^2 \theta}{2} + (-1)^k \omega_D \dot{f} \cosh 2f \cos \theta (2 + \sin^2 \theta) + \frac{\omega_0^2}{2} \sin^2 \theta - \frac{\omega_0^2 \beta_4}{4} \sin^4 \theta \sinh^2 2f \right]. \quad (34)$$

It can be seen that the imaginary component  $f = \text{Im } \phi$  of the instanton solution effects only on the real part of the Euclidean action. The system (33) is equivalent to a mechanical system with two degrees of freedom. Only one first integral is known for it:

$$\mathcal{E} = \frac{\hbar^2 N}{2Jz} \left[ \frac{\dot{\theta}^2 - \dot{f}^2 \sin^2 \theta}{2} - \frac{\omega_0^2}{2} \sin^2 \theta + \frac{\omega_0^2 \beta_4}{4} \sin^4 \theta \sinh^2 2f \right]. \quad (35)$$

Note that  $\mathcal{E} = 0$  for the separatrix solutions we are interested in. For this reason this system cannot be analyzed exactly. However, an approximate solution can be constructed in the physically interesting case, when  $\omega_D \ll \omega_0$ ,  $\beta_4 \ll 1$  and for any relation between  $\omega_D$  and  $\omega_0 \beta_4$ . In order to find such a solution, we note that Eq. (33a) from the system (33) is transformed into  $(\dot{\theta})^2 = \omega_0^2 \sin^2 \theta$  in the zeroth approximation in the small parameters  $\omega_D/\omega_0$  and  $\beta_4$ . In this case the constant solution  $f = f_0 = \text{const}$  satisfies Eq. (33b) and gives

$$\sinh 2f_0 = \frac{\omega_D}{\beta_4 \omega_0}. \quad (36)$$

It should be noted that the value of  $f_0$  is determined by the ratio of two small parameters and can be appreciable. Using this fact, we can write a refined equation for  $\theta(\tau)$

$$\ddot{\theta} = \omega_0^2 \sin \theta \cos \theta \left( 1 - \frac{\omega_D^2}{\beta_4 \omega_0^2} \sin^2 \theta \right). \quad (37)$$

The approximate solution constructed by us is valid if  $\dot{\theta} \simeq \omega_0 \sin \theta$ , i.e.

$$\omega_D^2 \ll \frac{\omega_0^2}{\beta_4}. \quad (38)$$

This condition may also hold in the case when the value of  $\sinh 2f_0 = \omega_D/\beta_4\omega_0$  is of order of unity, but it is still violated for  $\beta_4 \rightarrow 0$ . In this case the situation is similar to that observed for the domain wall dynamics:<sup>21</sup> the limit velocity of a domain wall in a tetragonal antiferromagnet with an odd axis with  $\phi \neq \text{const}$  tends to zero as  $\beta_4 \rightarrow 0$ , and no dynamic solution exists for  $\beta_4 = 0$ .

The value of the Euclidean action for the obtained solution is real and is defined by the expression

$$\mathcal{I} = \frac{\hbar^2 N \omega_0}{Jz} \left( 1 + \frac{\omega_D^2}{3\beta_4\omega_0^2} \right). \quad (39)$$

In the range of applicability of the constructed solution, i.e., when  $\omega_D$  and  $\beta_4$ , are small and when inequality (38) is satisfied, the inclusion of the DM interaction leads only to a small correction to the real part of the Euclidean action, the imaginary part being identically equal to zero.

For models of antiferromagnets with binary and hexagonal symmetry axes such an approximate solution cannot be constructed, but the analysis of these models is even simpler than in the case of antiferromagnets with a tetragonal symmetry axis. In these cases we can also verify that, if the exact solution  $\phi = \phi_k^{(0)} = 2\pi k/n$ ,  $\theta = \theta(\tau)$  does not exist, the solution has the same form as before:

$$\phi = \frac{2\pi k}{n} + if(\tau), \quad \theta = \theta(\tau), \quad (40)$$

and the term in the Lagrangian which is linear in  $d\mathbf{l}/d\tau$  contributes only to the real part of Euclidean action  $\mathcal{I}$ . It can be proved, however, that function  $f(\tau)$  is anti-symmetric and proportional to the parameter  $\omega_D/(\beta\omega_0)$ , which is always small (in contrast to the case of a tetragonal antiferromagnet, in which there appears the parameter  $\omega_D/(\beta_4\omega_0)$  whose value may be large). Consequently, we can seek the function  $f \ll 1$  using the same perturbation theory as for the domain wall dynamics in such antiferromagnets.<sup>21</sup> As a result, we obtain the following expression for the real part of the Euclidean action:

$$\mathcal{I} = \frac{\hbar^2 N \omega_0}{Jz} \left( 1 + \xi \frac{\omega_D^2}{\omega_0^2} \right), \quad (41)$$

where the numerical factor  $\xi$  is of order of unity. Thus, the correction to the result typical for the Lorentz-invariant model is always small. Note that no interference effects take place in this case. We arrive at the conclusion that the DM interaction effects on the tunnel probability in hexagonal and orthorhombic antiferromagnets.

## V. CONCLUDING REMARKS

The analysis of antiferromagnetic particles with a tetragonal symmetry axis shows that three possible types of the effect of the DM interaction on tunneling processes exist. The investigation of the remaining cases important for the analysis of crystalline antiferromagnets (orthorhombic or uniaxial with a hexagonal symmetry axis,

see the Table I) proved that these types include all possible cases for antiferromagnetic systems with a doubly degenerate ground state. In fact, all cases can be reduced to the following three types of behavior.

1. The principal axis is even, e.g.,  $4_z^{(+)}$  or  $6_z^{(+)}$ . In this case vector  $\mathbf{l}$  is real on all instanton trajectories and these trajectories are planar  $\phi = 2\pi k/n$ ,  $\theta = \theta(\tau)$ . The real part of the Euclidean action is independent of the DM interaction constant, the imaginary part is equal to zero, and destructive interference effects are absent. In this case tunneling can be described without taking into account the DM interaction.
2. The principal axis is odd, and there exists an exact real solution with the rotation of  $\mathbf{l}$  in the easy plane determined by the anisotropy. Such an example is a system of the type  $4_z^{(-)}2_x^{(-)}$ , in which the instanton trajectory is plane and the DM constant does not appear in the real part of the Euclidean action. In this case, however, the inclusion of the DM interaction leads to the emergence of the imaginary part of the Euclidean action and may effect on the tunneling probability due to the interference of instanton trajectories lying in different planes. Since the corresponding phase factor contains the large value  $N$ , tunneling can be suppressed completely due to the destructive interference even for small values of the DM constant.
3. The principal axis is odd, and the simple solution  $\phi = 2\pi k/n$  does not exist. In this case, the vector  $\mathbf{l}$  has both real and imaginary components, but all types of DM interaction change only the real part of the Euclidean action, this change being small in view of the smallness of the parameter  $d^2/(J\beta)$ . The imaginary part of the Euclidean action is equal to zero and destructive interference effects are absent.

Thus, the only important effect produced by the DM interaction is associated with the possibility of the interference of instanton trajectories in the case when their number is greater than two (an antiferromagnetic particle with the easy magnetization axis approximately corresponds to  $n > 2$ ). This effect can be observed for antiferromagnets with an odd principal axis in the case when the rotation of  $\mathbf{l}$  on the instanton trajectory also occurs through the odd axis. It is associated with the interference of pairs of instanton trajectories lying in different planes. Since  $\mathbf{l}$  is real in this case and all instanton trajectories are plane, an exact analysis can easily be carried out and the description of tunneling is reduced to the geometrical analysis described in the section III.

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  - <sup>39</sup> It should be noted that the situation in this case is basically different from the case of a particle with an uncompensated total spin  $S$ , where we have the potential of the field of the magnetic monopole for  $\mathcal{A}$  (see Eq. (4) above). This vector potential  $\mathcal{A}$  can be written only in a singular form with a singularity on the half-line emerging from the point of location of the monopole (the Dirac string). The total flux  $\Phi_{tot}$  through the sphere is equal to  $4\pi S$ , and, hence, the phase difference  $\Delta\Phi$  for two diametrically positioned trajectories is equal to  $2\pi S$ . For this reason, the phase factor  $\cos(\Delta\Phi/2) = 0$  for half-integral values of  $S$ , and tunneling is forbidden. It should also be noted that, having repeated Dirac's analysis concerning the uniqueness of the electron wave function is the monopole field, we can derive the condition  $\cos(\Phi_{tot}/2) = 0$  leading to the half-integer quantization of uncompensated spin (an example of the quantization of parameters; see Ref.<sup>35</sup>). In our case of an uncompensated antiferromagnetic particle in the presence of an external field and/or DM interaction,  $\Phi_{tot} = 0$  and, naturally, no quantization of parameters takes place.